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Feasibility of Fast Neutron Spectroscopy for Safeguards and Verification of Spent Fuel in Dry Cask Storage

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Abstract

Spent nuclear fuel is being stored in dry casks as an interim storage solution. This current research supports the development of a neutron spectroscopy system using helium-4 gas scintillation fast neutron detectors to convert the neutron spectra into quantifiable signatures. The goal is to use these signatures to identify diversion/removal of fuel and verify spent fuel parameters, such as cooling time and burnup. To validate this concept, Monte Carlo simulations were used to produce realistic neutron spectra from PWR assemblies inside a dry cask storage system. A detailed methodology is discussed for generating neutron spectra using the Next Generation Safeguards Initiative (NGSI) spent fuel libraries and a high-fidelity model of the HI-STORM 100S dry cask system. ORIGEN-S calculates the neutron emissions from the spent fuel rods and MCNP 6.1 performs the transport and shielding calculations. Various diversion scenarios were analyzed to determine the ability to detect missing assemblies around the periphery and center of the cask. Although a significant neutron flux is emitted from the cask, neutrons less than 1 MeV dominate the spectrum. There is limited feasibility of using the total and relative contributions of spontaneous fission and (α,n) to predict fuel parameters. However, simulations show the feasibility of using count rate statistics to identify diverted assemblies around the periphery of the cask. Fluxes outside the cask but nearby the missing assembly location are 6% to 8% lower when compared to fluxes from a fully loaded cask.

1 Introduction

The neutron and gamma source terms of spent fuel are dependent on parameters such as initial enrichment, burnup, and cooling time. The concentration of radioactive emitters varies as a function of the nuclide creation and destruction during burnup and radioactive decay over time [1]. The gamma spectra has been studied through multivariate analysis to predict spent fuel parameters [2], however the neutron spectra has had limited application and research for the same purpose. While many neutrons exiting the cask have interacted multiple times and provide limited usefulness for neutron spectroscopy, some higher energy neutrons are emitted with little or no scattering [3]. This study investigates the feasibility of measuring neutron count rates and neutron energy spectra for safeguards and verification of spent fuel in dry cask storage.

This current research supports the development of a neutron spectroscopy system using helium-4 gas scintillation fast neutron detectors, which have excellent gamma-insensitivity and neutron spectroscopy capability, to convert the neutron spectra into quantifiable signatures. The goal is to use these signatures to identify diversion/removal of fuel and predict spent fuel parameters, such as cooling time and burnup. This technique would satisfy the IAEA need for robust safeguards and verification technologies for ensuring the continuity of knowledge and integrity of radioactive materials stored within the dry casks. While current safeguards strategies for interim storage casks focus on containment and surveillance (C&S) techniques to maintain continuity of knowledge, this NDA technique would offer a supplemental method.

Simulating spent fuel inside a dry cask storage system is the first step to show a proof of concept for converting measured neutron count rates and energy spectra to quantifiable signatures. These simulations

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require generation of a neutron source term for the spent fuel rods and calculating the multiplication and shielding effect. This research combines a high-fidelity geometry for the HI-STORM 100S dry cask storage system with the Next Generation Safeguards Initiative (NGSI) spent fuel libraries [4]. The neutron source terms for the spent fuel assemblies were calculated using the ORIGEN-S module in the SCALE 6.1 system [5]. The multiplication and shielding analyses were performed in MCNP 6.1 developed by Los Alamos National Laboratory (LANL) [6].

It should be noted that the neutron energy spectra from spent fuel inside dry cask storage has been examined before. Smith et al. is one such study comparing experimental measurements to MCNP simulations of a loaded TN-32 dry shielded container [7]. Peerani et al. modeled PWR fuel assemblies inside a Castor V21/A canister [8]. We expand on those studies in several ways: (1) by simulating multiple assembly loadings with various parameters; and (2) by examining the fast neutron energy spectrum.

Key goals of this study include the following: (1) Is it possible to detect the diversion of a PWR fuel assembly from a HI-STORM 100S dry cask system by passive neutron measurements? (2) How is fast neutron spectroscopy affected by self-shielding from the assemblies and attenuation from the concrete overpack?

2 Computational Methodology

Generating the neutron spectra from spent fuel assemblies stored in dry cask storage is a multi-step process utilizing a variety of computational resources including the NGSI Spent Fuel Library, ORIGEN-S, and MCNP. Our previous work was limited to calculating transported neutron spectra from individual rods and assemblies [9]. This phase of the research adds the capability to calculate the transported neutron spectra from spent fuel assemblies inside dry cask storage.

The HI-STORM 100S cask system used in the simulations accommodates a wide variety of spent nuclear fuel assemblies and other radioactive materials in a single overpack using different multipurpose canisters (MPCs). The cask stores up to 32 PWR fuel assemblies or 68 BWR fuel assemblies in a vertical orientation. The overpack is a cylindrical container made from stainless steel and concrete. It provides structural and shielding protection for the MPC. The MPC is a stainless steel cylindrical canister with a honeycomb fuel basket that is loaded into the overpack in a vertical orientation. Neutrons are absorbed by Metamic, a boron carbide metal matrix composite material. Detailed dimensions and compositions of the HI-STORM 100 cask system can be found in the Final Safety Analysis Report from Holtec International [10].

The MCNP models of the HI-STORM 100S cask system are based on the geometry developed in the neutron fingerprinting research by Rauch [11]. Since our focus is on fast neutrons, all calculations used 200 energy groups with 0.10 MeV bins from 0 to 20 MeV. The MCNP model included the multipurpose canister (MPC), baseplate, pedestal, platform, inner shell, concrete shield, outer shell, lid top plate, lid shield block and radial plates. Assembly details included the guide/instrument tubes, gaps between fuel and cladding, Metamic, and basket steel. These assembly details provide a realistic basis for the transport and shielding calculations in MCNP. The helium-4 detectors were modeled as stainless-steel cylinders with an active length of 20 cm and an inner diameter of 4.4 cm for a total active volume of 304 cm^3. Figure 1 shows two cross sections of the MCNP geometry.

Simulations were performed with up to 32 assemblies inside of a HI-STORM 100S dry cask system. All ORIGEN-S calculations were performed on a conventional desktop computer running the Windows operating system. All MCNP simulations were performed on the University of Florida HiPerGator computing cluster. ORIGEN-S calculations were almost instantaneous. MCNP calculations took about two days for 10 billion neutron runs. Chen et al. note that the limiting factors in a complicated shielding problem are how realistic the calculation model can be [12]. Reliable source term estimation and high-fidelity geometry modeling are critical. ORIGEN-S was used to calculate a reliable source term estimate. The geometry used included appropriate details for accurate shielding and transport calculations.

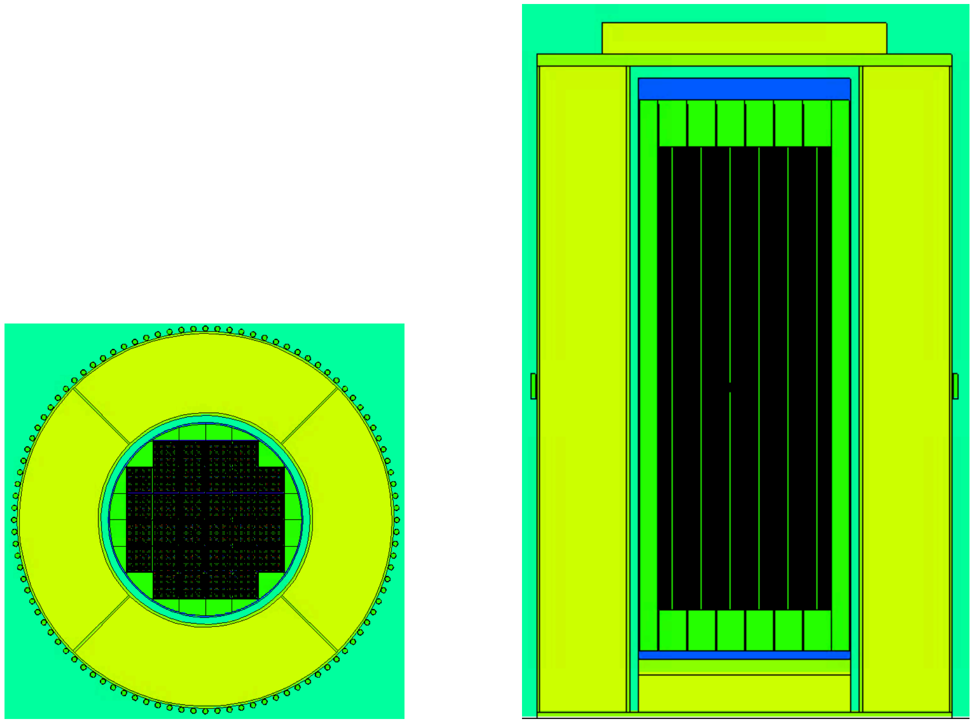

Figure 1. Horizontal and vertical cross sections of the MCNP model of the HI-STORM 100S cask system. Helium-4 detectors are spaced equally around the cask.

Initial cask loadings used assemblies with identical parameters from the NGSI Spent Fuel Library as the easiest scenario for detecting a diverted assembly. Additional loadings were developed based on real loading data from ORNL. Average surface flux tallies (F2) were used on the inside and outside cylindrical surfaces of the canister and overpack. Average cell flux tallies (F4) were used for the gas volume of each of the helium-4 gas scintillation detectors.

Unlike many simulations which homogenize the fuel, the fuel assembly structures are modeled explicitly. The NGSI spent fuel libraries have each rod as a single axial fuel region so it was not possible to calculate axial source term variation. Although the neutron energy spectra were calculated individually for each rod in the 17x17 PWR assemblies, there were only minor differences in the spectra. To simplify MCNP input files, the spectra for all 264 rods was averaged and set as the energy distribution for all rods in that assembly. This approach introduced a very minor approximation for the radiation transport.

Initial sensitivity analysis started with identical assemblies inside the HI-STORM 100 dry cask system. These had the same materials, neutron source strength, and neutron spectra emitted. The symmetry allows picking five different assembly diversion locations (1, 2, 6, 7, and 13) that are equivalent to removing any of the 32 assemblies as shown in Figure 2. Additional simulations were run with assemblies of varying average neutron source strengths to approximate real cask loadings. These asymmetric loadings were used to determine the effect on the neutron flux measured by the detectors in the same azimuthal area.

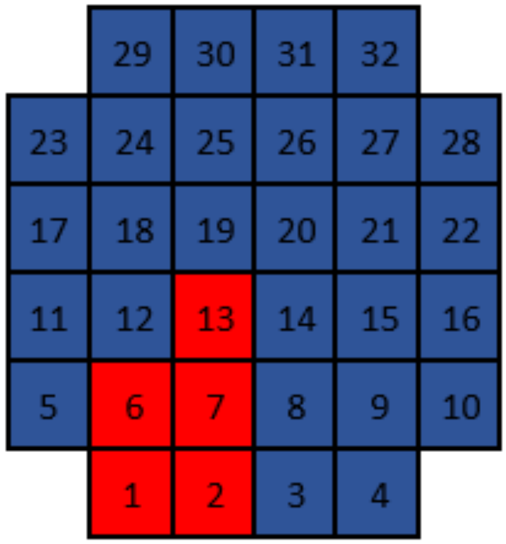

Figure 2. The loading pattern of 32 spent fuel assemblies in a HI-STORM 100S cask.

3 Results and Discussion

The sensitivity analysis results were completed for each of the diversion scenarios. Neutron fluxes were normalized per source neutron. Diverting an assembly results in less self-shielding of neighboring assemblies. Each of the 100 detector locations measures neutron fluxes around equal azimuthal angles around the cask. Figure 3 shows that outer fuel assemblies obscure the diversion of an inner fuel assembly. Detectors 71 through 80 are located close to the missing assembly locations. These detectors measure approximately 6% lower flux when assembly 1 is missing and approximately 8% lower flux when assembly 2 is missing compared to a fully loaded cask. When assembly 6, 7, or 13 is missing, the same detectors measure a flux within 2% of a fully loaded cask. The general size of the uncertainty is 1.5%. In addition, this is as close to an ideal scenario as possible with identical assemblies, neutron source strengths, and spectra. A real cask would have varying source strengths that would lead to more difficult analyses.

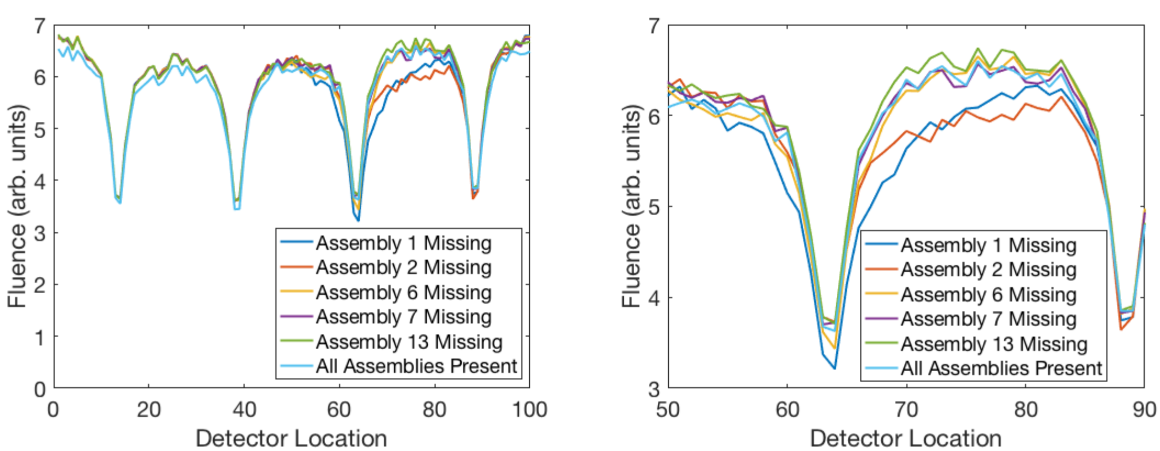

Figure 3. Fast neutron fluence at each detector from full cask and casks with assemblies removed from 5 different locations (left). Data subset zoomed into the area showing the fluence differences (right).

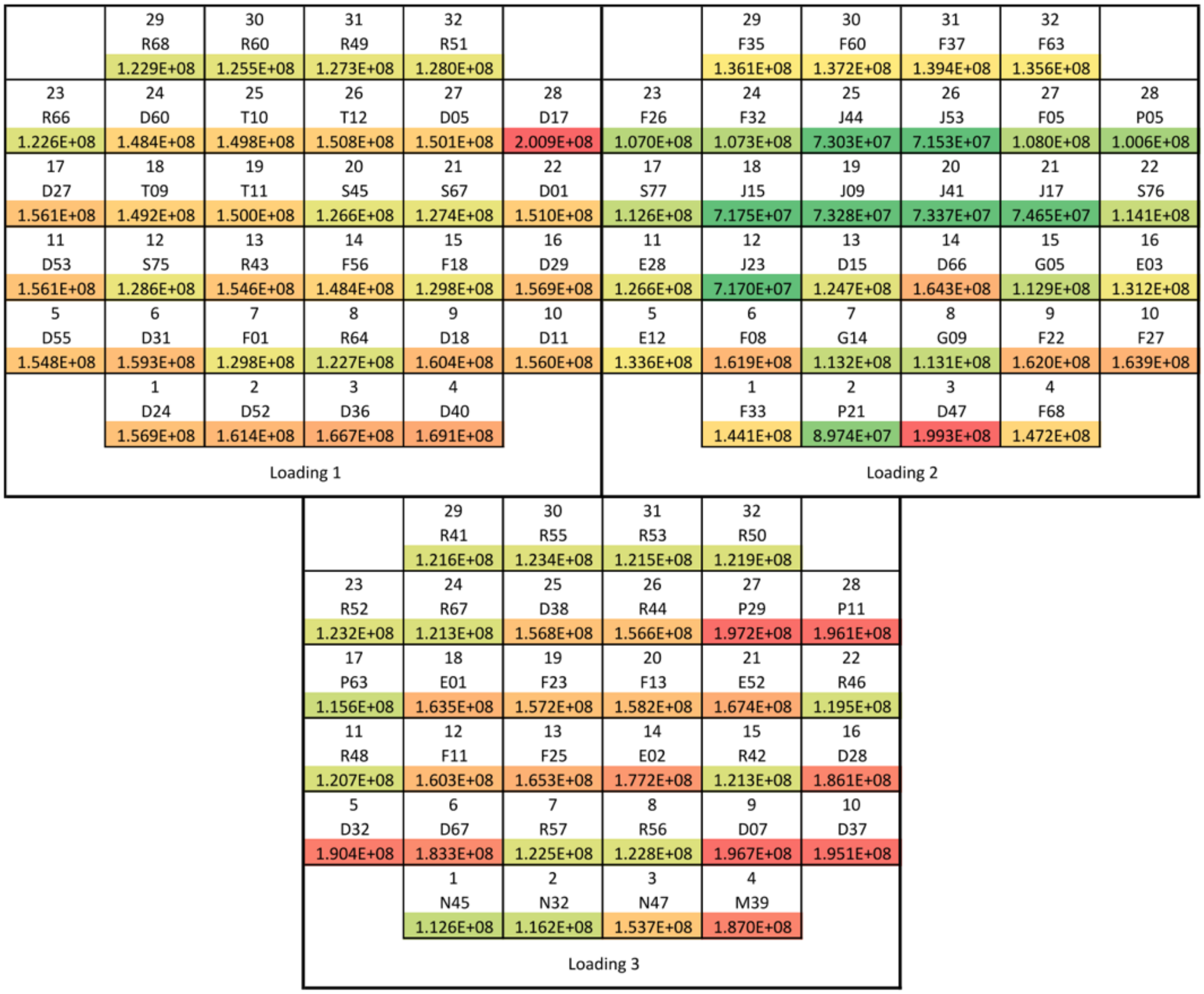

Figure 4. Loading pattern and average neutron source terms (n/s) per assembly for three real casks provided by ORNL [11].

The next set of simulations compared real cask loading data from ORNL to the identical assembly loadings. The loading pattern and average neutron source terms are shown in Figure 4. Figure 5 shows that the count rates measured by each detector are sensitive to the burnups of individual assemblies. Further investigations are needed to determine the extent to which an assembly with a weaker neutron source term may mimic a diverted assembly or an assembly with a stronger neutron source term may obscure a diverted assembly. Although a significant neutron flux exits the dry cask storage system, neutrons at lower energies dominate the spectrum. Over 90 percent of the neutrons are below 1 MeV. While higher energy neutrons are emitted, the reduced flux presents a challenge for obtaining reasonable statistics.

As our previous research showed, the (α,n) reaction neutrons have a maximum between 2.5 and 3.0 MeV. Our prior proposed technique estimated the number of spontaneous fission neutrons and (α,n) reaction neutrons based on the characteristics of their spectra: spontaneous fission neutrons peaked between 0.8 to 1 MeV and (α,n) reaction neutrons peaked between 2.5 and 3 MeV. This technique has limited applicability to spent fuel inside a dry storage cask with significant self-shielding and external shielding from the concrete. As shown in Figure 6, the self-shielding from the assemblies and concrete overpack results in mostly neutrons below 1 MeV emitted from the cask. The total neutron cross section of oxygen has a valley at approximately 2.3 MeV that results in a peak in the emitted neutron spectra.

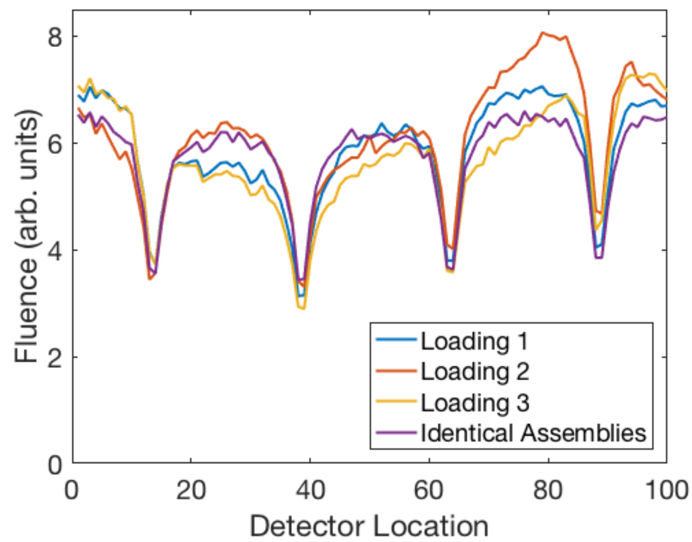

Figure 5. Neutron fluence at each detector for 3 different loadings compared to the loading with identical assemblies.

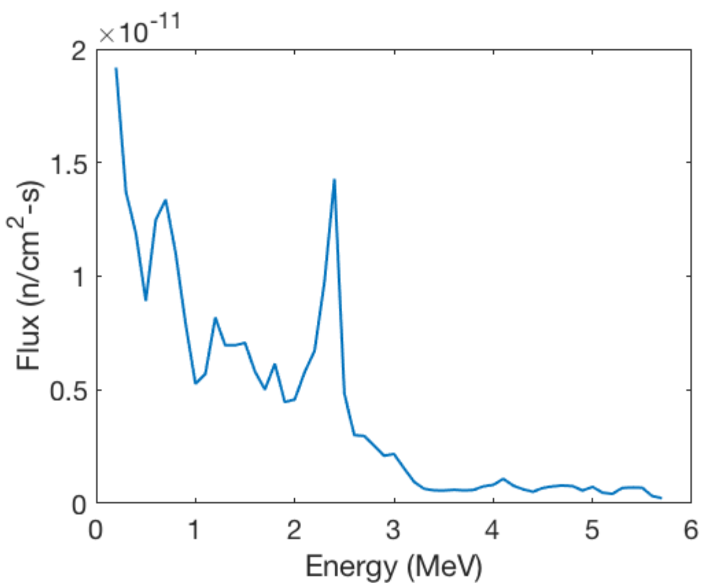

Figure 6. Surface flux tally (F2) on outside cylindrical surface of concrete overpack.

The tally on the MPC shows the neutron spectra with self-shielding from the assemblies but without the attenuation from the concrete overpack. Figure 7 shows that the neutron spectra inside the cask is still significantly affected by self-shielding from the assemblies. The difference between the internal and external neutron fluxes is approximately 5 orders of magnitude, illustrating the strong radiological shielding from the concrete overpack.

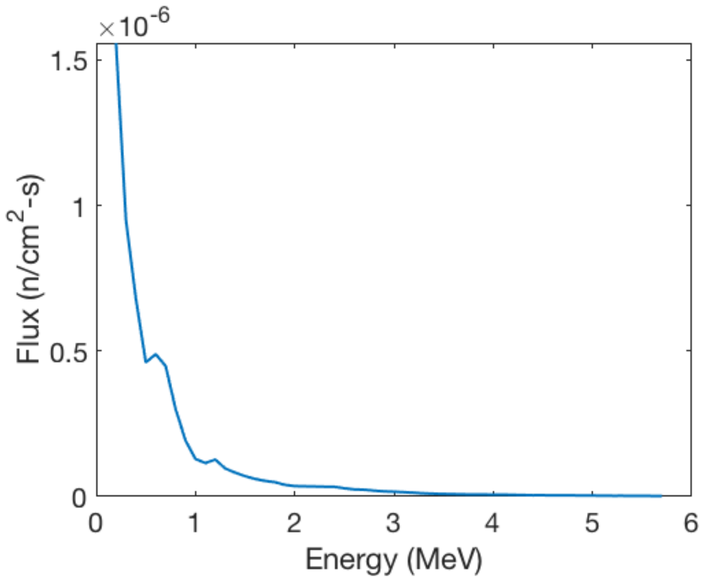

Figure 7. Surface flux tally (F2) on inside cylindrical surface of MPC.

The difficulty of identifying diverted inner assemblies is due to the self-shielding from the outer assemblies. Quantifying the contribution of each assembly to the overall neutron source term is the first step towards determining the sensitivity required to identify diversions. Figure 8 shows the contribution to the overall neutron source term from each assembly normalized to 1. There is a strong symmetry in the expected contribution for each assembly. Each assembly around the perimeter contribute slightly less than 5% of the total neutrons with about 2% of the neutrons coming from each assembly in the next layer inward. The innermost assemblies contribute only about 1% each to the total neutrons.

Figure 8. Contribution from each assembly to the total neutron flux on the outside cylindrical surface of the cask.

Based on the simulation results, fast neutron spectroscopy faces numerous challenges for spent fuel in dry cask storage due to self-shielding from the assemblies and attenuation from dry cask systems utilizing an overpack. Even spent fuel assemblies with different burnups, initial enrichments and cooling times have the same qualitive neutron energy spectra seen in Figures 7 and 8. These results show that neutron count rates are feasible for identifying diverted assemblies.

The reliability of the tally results was measured by the relative error for each energy group, overall relative error and the results of the ten statistical tests built into MCNP. The uncertainties in these simulations come from several sources: uncertainties in the nuclear data, geometry model, and neutron source simplifications. These results are obtained using the latest evaluated nuclear cross section data in ENDF/B-VII. Relative error increased substantially in higher energy groups (>8 MeV). There were not enough neutrons passing the surface at these energies to generate reasonable statistics. Neutrons at those energies would be of negligible impact since they would have a reduced probability of detection.

Computing resources limit the statistical accuracy of the simulations. The source term generation in ORIGEN is not computationally intensive, however the transport and shielding calculations in MCNP require significant computational resources. Researchers had to balance increasing the number of neutrons in each simulation to obtain better statistics or having more simulations to model different cask loading scenarios.

4 Summary and Conclusions

The methodologies and results from this study should improve the usefulness of future analyses of spent fuel in dry cask storage. Future computational efforts include developing a mathematical model to predict count rates from cask loading information. Important effects to incorporate include self-shielding from assemblies and significant attenuation from the dry cask system itself.

Fast neutron spectroscopy for safeguards and verification of spent fuel in dry cask storage is feasible. Computational simulations have shown the ability to identify diverted assemblies on the outside perimeter. However, diversion of interior assemblies leads to very weak changes to count rates. All neutron energy spectra measured on the outside of the cask appear qualitatively similar due to self-shielding from the assemblies and attenuation from the concrete overpack. This offers minimal, if any, opportunity to predict spent fuel parameters based on the neutron energy spectra. Future efforts include neutron spectroscopy of dry casks using helium-4 scintillators in a prototype system.

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